REMOVAL OF TAR IN PRODUCER GAS BY CLARIFYING THE COMBUSTION MECHANISM OF THE INVERSE DIFFUSION FLAME

Noraki NAKATSUKA*, Miki TANIGUCHI**, Jun HAYASHI*, Kenichi SASAUCHI**, Mayumi MATSUDA*** and Fumiteru AKAMATSU*

*Dept. of Mech. Eng., Osaka University, 2-1, Yamada-oka, Suita, Osaka 565-0871, Japan
**Chugai Ro Co., Ltd., 2-4, Chikko-shinmachi, Nishi-ku, Sakai, Osaka 592-8331, Japan
***J-Science Lab Co., Ltd., 3-1, Hiuchigata-cho, Kamitoba, Minami-ku, Kyoto 601-8144, Japan

ABSTRACT Experimental study for clarifying the effect of combustion mechanism on reforming producer gas was conducted. The inverted diffusion flame was formed in the actual gas reformer by partial combustion of producer gas. Direct observation and laser diagnosis were applied to the inverse diffusion flame. Experimental parameters are steam concentration in supplied model producer gas ($X_{steam}$) corresponding to moisture content of wood. The main results are as follows. In the condition of 20% concentration of oxygen, flames at nozzle and lifted flames were formed in the same supply condition by hysteresis. When the flames at nozzle were formed, the exergy yield was increased with the increase of $X_{steam}$ because soot formation was suppressed and steam reforming of model tar could proceed. In contrast, when the lifted flames were formed, the exergy yield was decreased with the increase of $X_{steam}$ because of formation of SOF (soluble organic fraction).

Keywords: Inverse Diffusion Flame, Tar, Biomass, Soot, Exergy, Gas Reformer

1. INTRODUCTION

In recent days, energy use of woody biomass, which is not competing with food on the land use, gathers intensive attention. Woody biomass should be converted into energy by small energy conversion system because of its small energy density. As this small energy conversion system of woody biomass, we focus attention on the electric generation by woody biomass gasification [1]. Generally speaking, formation of tar in producer gas is crucial problem in case of woody biomass gasification. Tar is complex substance consisted of many kinds of hydrocarbons. When the producer gas that contains tar is cooled before supplied to gas engine for the electric generation, tar is condensed to clog the pipes and erode the inside of the gas engine. Therefore, tar concentration in the producer gas needs to be decreased. In this circumstance, it is considered as one of the effective ways to decrease the tar contained in the producer gas that partial combustion of producer gas for oxidative and thermal cracking of tar in the gas reformer equipped at the subsequent stage of biomass gasifier [2].

In the actual gas reformer, inverse diffusion flame is formed by supplying oxidizer in the high-temperature producer gas. Cracking and polymerization of tar occur simultaneously at the proximity of the flame. Cracked tar is converted into useful fuel gases and carbon dioxide. On the other hand, polymerized tar could grow to soot through polycyclic aromatic hydrocarbons (PAH) [3]. Soot formation is a significant problem of the gas reformer because of decrease of energy efficiency and increase of complexity of the gas reformer by installing soot-trapping filters. In past researches [4], however, mechanisms of cracking and polymerization of tar at the proximity of the inverse diffusion flame remain to be completely investigated because of complexity of supplying method of oxidizer and difficulty of applying the soot measurement method to the inverse diffusion flame formed in high-temperature fuel atmosphere.

Therefore, it is the final goal of the present research that the mechanisms of cracking and polymerization of tar at the proximity of the inverse diffusion flame are clarified. In order to achieve this final goal, experimental apparatus that enables to apply the laser diagnostics to the inverse diffusion flame and the purpose-built system gas chromatograph are newly developed.

2. EXPERIMENTAL

2.1 Supplying System and Combustion Chamber

![Fig. 1 Schematic of the experimental apparatus.](image-url)
Figure 1 shows the schematic of experimental apparatus. As model producer gas, mixed gas of hydrogen, methane, carbon monoxide, carbon dioxide, steam (water vapor) and toluene vapor is supplied from supplying system. Volume flow rate of each gas compound except of steam and toluene is controlled by the mass flow control valve. Steam corresponding to moisture content of biomass is supplied as water by water pomp and evaporated by primary heater. Toluene as a model tar compound is supplied to hot gas mixture by syringe with micro feeder. Temperature of model producer gas at TC2 is controlled at 500 °C by secondary heater. An inverse diffusion flame is formed in combustion chamber as shown in Fig. 1. Cross section of gas passage is 80 mm square.

Oxidizer is supplied from the under part of the experimental apparatus and introduced to the ignition point by straight stainless nozzle penetrating the combustion chamber concentrically. The outer diameter of stainless pipe is 8 mm and the inner diameter is 6 mm. The inverse diffusion flame is formed downstream of the stainless pipe for oxidizer supply in the model producer gas heated up to the auto-ignition temperature. The inverse diffusion flame is observed through the double quartz windows forming three sides of the combustion chamber. Owing to the double quartz windows, laser diagnostics described later in detail can be applied.

The burnt model producer gas sampled at the point shown as “Gas sample” in Fig. 1 is analyzed by the newly developed system gas chromatograph described later in detail. The burnt gas after sampling is dewatered by the drain line and completely burned by flare stack.

2.2 Laser Diagnostics

There are few researches that focus on soot formation in partial combustion of producer gas contains tar [4, 5]. Furthermore, there are no researches in which the laser diagnostics is applied to the inverse diffusion flames formed in high-temperature or tar-containing fuel atmosphere. Soot formation at the proximity of the flame can be observed as luminous flame. However, the flame luminescence intensity observed by direct observation contains line-of-sight information. Furthermore, the flame luminescence intensity itself weaves luminescence of soot with chemiluminescence from the flame. In this study, laser induced incandescence (LII) is applied to the sooting inverse diffusion flames in order to measure soot volume fraction without alternation in the flame structure and effect of chemiluminescence from the flame. Figure 2 shows schematic of the optical system of LII.

![Schematic of the optical system of LII](image)

In this study, a second harmonic (532 nm) of the Nd:YAG laser (Quanta-Ray, Spectra Physics Inc.) is used. The repetition frequency of laser is set at 10 Hz, the duration time is approximately 8 ns, and the energy is approximately 34 mJ/pulse. Three cylindrical lenses are used to form a laser sheet that is 30 mm in height and 500 μm in width. Therefore, the average laser fluence per pulse is 0.23 J/cm² and the mean laser flux is 2.8 × 10⁷ W/cm². The laser sheet is aligned vertically with the central axis of the oxidizer supply nozzle and measurement region is from 70 mm to 100 mm above the oxidizer supply nozzle exit.

A high speed digital camera (Phantom Ver. 5.0, Vision Research Inc.) with an image intensifier (C4273MOD, Hamamatsu Photonics Inc.) and a 50-mm focal length, f/1.2 lens (Nikkor, Nikon Inc.) is used for LII signal detection. The camera is oriented perpendicular to the laser sheet. Measurements of LII are conducted with the band-path optical interference filter (center wavelength: 400 nm, half band width: 60 nm) to collect LII signal efficiently. The gate width of the image intensifier is set to 70 ns to collect maximum LII signal without chemiluminescence from the flame. The digital camera and the image intensifier are synchronized with the Nd:YAG laser by pulse generator. Timing diagram of the LII measurement system is shown in Fig. 3.

![Timing diagram of the LII measurement](image)

**Fig. 3** Timing diagram of the LII measurement.

### 2.3 Multicomponent Burnt Gas Analyzing System

In order to figure out the energy balance or the material balance involving tar component accurately, main components of producer gas have to be analyzed by the same gas analyzing system. Because of multicomponent of partially-combusted model producer gas, purpose-built online system gas chromatograph with parallel three detectors (one TCD and two FIDs) is newly-developed. Furthermore, partially-combusted model producer gas should be sampled with kept the temperature high because it contains many condensable components such as steam, toluene, phenol and naphthalene. Though there are few researches in which the online system gas chromatograph [6, 7] is introduced, there have been no system gas chromatograph that can analyze condensable components including steam and phenols with permanent gases simultaneously. Figure 4 shows schematic of the newly-developed system gas chromatograph.

At the heated pipe (HP*) inlet, silica wool is packed to remove particulate matter of burnt gas before quantification of the system gas chromatograph. The temperature of heated pipe is kept at 300 °C at HP* to prevent condensable components from condensation. The first online gas chromatograph equipped with thermal conductivity detector (TCD) can analyze hydrogen, methane, carbon monoxide, carbon dioxide and steam
At first, hydrogen, methane and carbon monoxide are separated on AC1 (packed column), and then carbon dioxide and steam are separated on AC2 (packed column).

The second online gas chromatograph equipped with flame ionization detector (FID) can analyze PAH (2 ~ 5 rings) and phenolic components (System II). PAH and phenolic components are separated on AC3 (capillary column) with programmed temperature.

The third online gas chromatograph equipped with flame ionization detector (FID) can analyze from C2 to C4 components e.g. ethane and acetylene, and benzene, toluene, xylenes (BTX) and phenylacetylene (System III). At first, from C2 to C4 components are separated on AC4 (packed column). Then, xylenes and phenylacetylene are separated on AC5 (packed column). At last, toluene, supplied as model tar compound, with high concentration is separated on AC6 (packed column).

The composition of producer gas based on that of producer gas derived from woody biomass gasified by the rotary kiln gasifier at 600 °C. Lop in Table 1 represents liter at 25 °C, 1 bar based on thermodynamic property of gases.

<table>
<thead>
<tr>
<th></th>
<th>0 wet%</th>
<th>10 wet%</th>
<th>20 wet%</th>
<th>30 wet%</th>
<th>40 wet%</th>
</tr>
</thead>
<tbody>
<tr>
<td>H2</td>
<td>1.60</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>CH4</td>
<td>2.40</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO</td>
<td>4.00</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO2</td>
<td>2.00</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C2H6</td>
<td>0.46</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H2O</td>
<td>17.40</td>
<td>13.05</td>
<td>8.70</td>
<td>4.35</td>
<td>0</td>
</tr>
<tr>
<td>N2</td>
<td>17.40</td>
<td>13.05</td>
<td>8.70</td>
<td>4.35</td>
<td>0</td>
</tr>
</tbody>
</table>

Steam concentration in supplied model producer gas (Xsteam) of the condition of 40 wet% is corresponding to moisture content of raw cedar wood. Toluene (C7H8) is supplied as model tar compound. Tar consists of many kinds of hydrocarbons and persistent compounds in tar could be aromatic hydrocarbons derived from lignin constructing woody biomass [8]. Because the aromatic hydrocarbons derived from lignin could path through benzene, toluene and xylenes polymerized to form PAH [3], toluene is one of most appropriate model compound of tar for investigating the cracking and polymerization mechanism of persistent tar. In each steam volume flow rate condition, nitrogen is supplied as shown in Table 1 to equalize the total volume flow rate of model producer gas. Cross-section average flow velocity of model producer gas at 500 °C is approximately 0.19 m/s.

As oxidizer, mixed gas of 20 % of oxygen and 80 % of nitrogen (volume fraction) is supplied from the straight stainless nozzle with 8 mm outer diameter and 6 mm inner diameter. Total flow rate of oxidizer is 5.7 Lop/min, cross-section average flow velocity of oxidizer at 25 °C is 3.4 m/s and Reynolds number when nozzle inner diameter is adopted as characteristic length is 1331.

### 3. RESULTS AND DISCUSSION

#### 3.1 Soot Formation

Figure 5 shows direct photographs of the inverse diffusion flames. Direct photographs of flames at nozzle are shown in Fig. 5 (a) and those of lifted flames, which are lifted from nozzle rim of oxidizer supply nozzle, are shown in Fig. 5 (b).

In this study, flames at nozzle and lifted flames were formed in the exactly same condition including flow velocity of oxidizer. It could be considered that there is a hysteresis in flame formation. The flame structure is determined by this hysteresis. Technically speaking, flames at nozzle were formed by decreasing in oxygen concentration from 40 %, at which only flames at nozzle could be formed, to 20 %, Lifted flames were formed by supply oxidizer with 20 % oxygen concentration from the beginning.

As shown in Fig. 5, luminescence intensity of luminous flame of flame at nozzle was much more intense than that of lifted flame in all Xsteam % condition. In addition, luminescence intensity of luminous flame became weakened with the increase of Xsteam in each flame structure. Then, LII measurement is applied to flames at nozzle to investigate the effect of Xsteam on soot volume fraction.

![Schematic of the system gas chromatograph](image)

Fig. 4 Schematic of the system gas chromatograph.

### 2.4 Experimental Condition

Table 1 shows the composition of model producer gas in Lop/min. The bottom line represents the tip of oxidizer supply nozzle and lines are 10 mm intervals.

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Fig. 6 Two-dimensional distribution of soot volume fraction in flames at nozzle detected by laser induced incandescence. White color represents LII signal and intensity of LII signal is approximately proportional to soot volume fraction.

Figure 6 shows two-dimensional distribution of soot volume fraction. As shown in Fig. 6, soot volume fraction was decreased with the increase of \( X_{\text{steam}} \). This result agrees with tendency of luminescence intensity of luminous flame (see Fig. 5 (a)). As for lifted flames, soot volume fraction in all \( X_{\text{steam}} \) condition was as low as flame at nozzle in condition of 40 wet%. Here, soot formation could be critically influenced by flame temperature. Table 2 shows temperature of the inverse diffusion flames in all condition without correction of radiation heat. Flame temperature was measured by the B-type thermocouple with 500 \( \mu \)m wire diameter. Measurement point is 75 mm above the nozzle exit and on the central axis of the nozzle.

Table 2 Flame temperature of each flame structure in °C.

<table>
<thead>
<tr>
<th></th>
<th>0 wet%</th>
<th>10 wet%</th>
<th>20 wet%</th>
<th>30 wet%</th>
<th>40 wet%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flames at nozzle</td>
<td>1047</td>
<td>1104</td>
<td>1114</td>
<td>1102</td>
<td>1074</td>
</tr>
<tr>
<td>Lifted flames</td>
<td>1088</td>
<td>1112</td>
<td>1093</td>
<td>1083</td>
<td>1090</td>
</tr>
</tbody>
</table>

As shown in Table 2, there was not so large difference in flame temperature between the flame structure and \( X_{\text{steam}} \). Therefore, suppression of soot formation by the increase of \( X_{\text{steam}} \) could be owed to change in chemical reaction and combustion state. In addition, flame structure was also considerably influential on soot formation.

3.2 Exergy Yield

In general, the cold gas efficiency depending on enthalpy analysis is often used in biomass gasification field as an indicator for energy efficiency estimation. However, the available maximum theoretical work (i.e. exergy) is different from each component that possesses the same enthalpy because standard entropy is different for each component. In this study, therefore, exergy yield is used as more practical indicator. Distribution of exergy yield is defined as follows.

\[
\text{Distribution of exergy yield, } (\%) = \frac{E_{\text{b, i}}}{E_a} \times 100 \quad (1)
\]

Where \( E_{\text{b, i}} \) is each exergy at 300 °C of major components in burnt model producer gas (hydrogen, methane, carbon monoxide, \( C_2 \) compounds, benzene, toluene, phenol and naphthalene). \( E_a \) is total exergy at 300 °C of unburnt model producer gas. Figure 7 (a) and (b) shows distribution of exergy yield as functions of steam concentration in supplied model producer gas (\( X_{\text{steam}} \)) in the case of flames at nozzle and that of lifted flames, respectively.

Fig. 7 Distribution of exergy yield as functions of steam concentration in supplied model producer gas.

As shown in Fig. 7 (a), effective exergy yield (available for gas engine) that is exergy yield except for exergy of condensable components such as toluene was increased with the increase of \( X_{\text{steam}} \) in the condition from 20 wet% to 40 wet%. In addition, in the condition from 20 wet% to 40 wet%, total exergy yield was increased with the increase of \( X_{\text{steam}} \). This result could be explained as follows: soot formation was suppressed and steam reforming of toluene represented as one-step overall reaction (shown in Eq. (2)) proceeded with the increase of \( X_{\text{steam}} \)

When soot formation was suppressed, exergy yield was increased because exergy of soot that could not be analyzed by the system gas chromatograph and exergy loss due to radiation heat by luminescence of soot were decreased. When steam reforming of toluene proceeded, exergy of products was increased because the reaction is endothermic reaction. Then, carbon monoxide and hydrogen are products of this reaction. In the condition from 20 wet% to 40 wet%, distributions of exergy yield of carbon monoxide and hydrogen were increased with the increase of \( X_{\text{steam}} \). This could be one of the evidences for proceeding of the reaction.

- Steam reforming of toluene
\[
C_7H_8 + 7H_2O \rightarrow 7CO + 11H_2 \quad \Delta E_{300 \circ C} = 216 \text{ kJ/mol} \quad (2)
\]

Meanwhile, in the condition from 20 wet% to 40 wet%, total exergy yield was decreased with the increase of \( X_{\text{steam}} \) in the case of lifted flames as shown in Fig. 7 (b).
This is exactly the opposite tendency compared with the case of flames at nozzle. As for details, in the condition from 20 wet% to 40 wet%, distribution of exergy yield of carbon monoxide was decreased and that of hydrogen was not increased as much as in the case of flames at nozzle with the increase of \( X_{\text{steam}} \). This result could indicate that the decrease of toluene was not due to steam reforming of toluene. Furthermore, soot formation in the case of lifted flames was much lower than in the case of flames at nozzle.

Then, carbon yield should be used in order to investigate what product is derived from toluene that was not reformed by steam reforming.

### 3.3 Carbon Yield

Distribution of carbon yield is defined as follows.

\[
\text{Distribution of carbon yield, (\%)} = \frac{C_{\text{b,i}}}{C_{\text{a}}} \times 100 \tag{3}
\]

Where \( C_{\text{b,i}} \) is each carbon mole number of major components in burnt model producer gas (hydrogen, methane, carbon monoxide, \( C_2 \) compounds, benzene, toluene, phenol and naphthalene). \( C_{\text{a}} \) is total carbon mole number of unburnt model producer gas.

Figure 8 (a) and (b) shows distribution of carbon yield as functions of \( X_{\text{steam}} \) in the case of flames at nozzle and that of lifted flames, respectively. As shown in Fig. 8 (a), total carbon yield was approximately constant with \( X_{\text{steam}} \) in the case of flames at nozzle. Meanwhile, in the condition from 20 wet% to 40 wet%, total carbon yield was decreased with the increase of \( X_{\text{steam}} \) in the case of lifted flames as shown in Fig. 8 (b). From these results, the decrease of exergy yield in the condition from 20 wet% to 40 wet% could be caused by formation of products contains carbon atom. Here, in case of lifted flames, more particulate matter (PM) was generated and deposited than in case of flames at nozzle.

Model producer gas is diluted by steam, carbon dioxide and nitrogen same as mixture supplied to the internal combustion engine with exhaust gas recirculation (EGR). The EGR is low-temperature combustion method for reduction of fuel consumption rate in partial load condition and emission of nitrogen oxide. In recent days, smokeless rich combustion that is simultaneous reduction method of soot and nitrogen oxide in the condition of fuel rich and large amount of EGR has been investigated for the diesel engine. In smokeless rich combustion condition, soluble organic fraction (SOF) that contains PAH is formed because the reactions from PAH to soot are suppressed [9].

Figure 9 (a) and (b) shows schematic of flame structure of the flames at nozzle and the lifted flames, respectively. As shown in Fig. 9 (a), the flames at nozzle are approximately bulk diffusion flame. Meanwhile, base of the lifted flames could have premixed flame segment because fuel and oxidizer mix immediately downstream of the nozzle rim, creating a mixture with a composition ranging from fuel lean to fuel rich. Eventually, the rich premixed flame segment whose combustion configuration is similar to smokeless rich combustion could be formed at the base of the lifted flame. That is to say, it could be considered that SOF was formed here. Thus, the decrease of exergy yield and carbon yield in the case of lifted flames could be due to formation of SOF at the flame base.
4. CONCLUSION

Partial combustion method could be effective way to reform tar in producer gas in the gas reformer equipped at subsequent stage of biomass gasifier. In this study, effects of flame structure and steam concentration in producer gas on polymerization of model tar compound and soot formation were investigated by use of the experimental apparatus in which the laser diagnostics can be applied to the inverse diffusion flames. The results obtained by the experimental consideration are as follows.

(a) In the case of flames at nozzle, total exergy yield was increased with the increase of steam concentration in supplied model producer gas ($X_{\text{steam}}$) in the condition from 20 wet% to 40 wet%. This could be explained because soot formation was suppressed and steam reforming of toluene that is endothermic reaction proceeded with the increase of $X_{\text{steam}}$.

(b) In case of the lifted flames, total exergy yield was decreased with the increase of $X_{\text{steam}}$ in the condition from 20 wet% to 40 wet%. This could be explained because soluble organic fraction (SOF) that consisted mainly of polycyclic aromatic hydrocarbons (PAH) was formed more with the increase of $X_{\text{steam}}$ at the flame base than in case of the flames at nozzle.

(c) The flames at nozzle could be suitable flame structure compared with the lifted flames, and higher effective exergy yield could be obtained in high $X_{\text{steam}}$ condition. Because soot formation was suppressed and effective exergy yield was increased with the increase of $X_{\text{steam}}$, flame needs to be stabilized in order to prevent the flames at nozzle from transition to the lifted flames.

5. REFERENCES